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## Abstract[1]

The electron capture cross sections of hydrogen cluster ions $H_n^+$ colliding with atomic helium have been measured in a large range of cluster size ( $5 \leq n \leq 35$ ) for the same velocity (1.5 $v_0$ , 60 keV/u). While the electron capture cross section decreases from the $H^+$ ion to the $H_3^+$ one, the cluster electron capture cross section is found to be independent of the cluster size and nearly equal to the capture cross section of the $H_3^+$ ion. The electron capture by hydrogen clusters on a helium atom is a process involving only the $H_3^+$ core of the cluster where the positive charge is localized. It appears that this very localized electron capture is not disturbed by the presence of molecules, up to sixteen, around the $H_3^+$ core.

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## I. INTRODUCTION

Hydrogen is by far the most abundant element in the Universe and molecular hydrogen $H_2$ is known to dominate in cool regions. Otherwise, as observed in Jupiter atmosphere, $H_3^+$ is supposed to have an important role in the interstellar medium as an initiator of chains of chemical reactions [1]. Recent quantum Monte Carlo simulation [2] and quantum chemical calculations [3] have investigated the effect of protonation of pure hydrogen clusters $(H_2)_n$ at low temperature. It was shown that the added proton gets trapped as a very localized $H_3^+$ impurity in the cluster and is surrounded by stable shells of solvating $H_2$ molecules. In recent years, research in cluster physics has expanded from the study of the isolated species in the gas phase to the interactions of atomic or molecular clusters with atoms, molecules and others clusters [4]. When a beam of molecular or cluster ions collides with a gas target, there occur several competing reactions involving dissociation, electron capture, ionization, etc. In particular, electron capture processes in ion-atom collisions play an important role in astrophysics, atmospheric physics and plasma physics. Then, studies of electron-capture cross sections by protons [5] and molecular hydrogen ions on different targets [6] have been pursued by many investigators due also to the inherent importance of this fundamental process. In this paper, we deal with the collisional interaction of the $H_n^+$ mass-selected cluster ions with helium atom where various types of elastic, inelastic, and charge transfer processes may occur.

To our knowledge, no result on cluster electron capture is available for various cluster sizes at the same velocity. Low-velocity collisions of $K_n^+$ and $Na_n^+$ cluster ions of different sizes with a Cesium vapor have been studied at a fixed energy laboratory (few keV)[7]. Total charge-exchange cross sections are measured and the resulting neutral products are shown to conserve the parent mass or evaporate at most either one atom or dimer. Especially, cross sections for $Na_n^+$ clusters ranging from the monomer to the 21-mer lie between 40 to 10 Å$^2$, decreasing slowly with increasing size. Nevertheless, in this experiment, the electron capture cross sections are shown to exhibit a strong dependence over cluster velocity [7]. Collisions involving fullerene ions at collision energies varying from few eV to 100 keV in the laboratory frame have also been studied [8-10]. The electron-capture cross sections are measured for fullerenes and rare gas atoms targets. Recently, capture cross section of 10 MeV $C_5^+$ clusters colliding with helium atoms have been measured [11].

In the present work, we report on a study of charge-exchange between mass-selected swift hydrogen clusters ions $H_n^+$ and atomic helium,

$$H_n^+ + He \longrightarrow H_n^{\cdot} + He^+,$$

in a large size range ( $3 \leq n \leq 35$ ) at the same velocity of 1.55 $v_0$ (60 keV/u), where $v_0$ is the Bohr velocity (fast cluster-ion/atom collision regime).

## II. EXPERIMENT

Experiments have been performed at the cluster facility of the Institut de Physique Nucléaire de Lyon [12]. The schematic experimental set up (Fig. 1) has been described in detail elsewhere [13]. Briefly, swift mass-selected ionized hydrogen clusters $H_n^+$ are produced and sent to the collision chamber. Before entering the chamber, the beam is collimated by two apertures giving an angular divergence equal to 0.16 mrad. The beam then crosses a gaseous jet formed into a vertical cylindrical capillary tube. Measurements at various input pressures are made in order to check the single-collision conditions. According to previous studies on the target thickness [14], absolute cross sections are deduced from the measurements. A magnetic analyser is then used to deflect the $H_p^{q+}$ ions according to the q/p ratio. Those are either no-dissociated clusters (p=n) or fragments resulting from dissociation (p<n). Only neutral or singly charged fragments (q=-1, 0 or +1) are observed. The detection is made with silicon solid-state detectors intercepting the various q/p trajectories.

However, the neutral fragments are not analysed with respect to their mass. For each dissociated cluster, the information given by the detection of the neutral fragments is the sum of their masses. A typical example ($H_{13}^+$ incident clusters) is given in Figure 2a. This spectrum has been obtained for a given number of incident clusters (~1.1 $10^5$) and for a target thickness of $1.41 \times 10^{14}$ at/cm$^2$ for which the single-collision conditions are fulfilled (20 % of dissociated clusters). It shows 13 separate peaks and the number $S_N$ associated with each peak corresponds to the value of the sum of the mass numbers of all the neutral fragments coming from the dissociation of a cluster. In this paper, we deal with the last peak ($S_N$ = 13) which corresponds to the following fragmentation channel where all the fragments are neutral :

$$H_{13}^+ + He \longrightarrow \Sigma\mu_i H_i + He^+, \text{ with } \Sigma(\mu_i\, i) = 13.$$

where $\mu_i$ is the number of fragments $H_i$.

This channel corresponds to the capture of a target electron by the $H_{13}^+$ cluster ion. The electron capture is followed by the dissociation of the excited neutral cluster produced. Indeed, measurements of the angular distributions of the neutral fragments have been performed and a first analysis of these data indicates that neutral fragments are mainly atomic and molecular hydrogen.

Figure 2b displays the spectrum obtained without helium gas target for the same number of incident clusters as in Figure 2a. These events are due to collisions with the residual gas. In the last peak corresponding to the electron capture process, the number of these spurious events corresponds to 8% of the peak ($S_N = 13$) in Figure 2a (about 0,07 % of the number of incident clusters). These spectra (Fig. 2a and Fig. 2b) have been measured for all the cluster sizes varying from 5 to 35, odd values, and also for the $H_2^+$ and $H_3^+$ molecular ions.

## III. DATA ANALYSIS

The absolute electron capture cross section can be deduced from the measurements by two different methods, the growth-rate method and the branching-ratio method. The use of these two methods could provide an evaluation of the uncertainty over the measured cross sections.The growth-rate method consists in the measurement of the number of events in the peak associated to electron capture ($S_N$=n) in the spectrum of detection of the neutral fragments for various target thicknesses $\varepsilon$ and for a given number of incident clusters. Then, we deduce the fraction $F_c^n(\varepsilon)$ of electron capture events versus the target thickness $\varepsilon$. In a single-collision regime, the fraction depends on the target thickness as follows

$$F_c^n(\varepsilon) = F_c^n(0) + \sigma_c^n \varepsilon \qquad (1)$$

where $\sigma_c^n$ is the electron capture cross section and $F_c^n(0)$ corresponds to the fraction of electron capture events without gas target (interaction with the residual gas).

As shown in Figure 3, for the incident $H_3^+$ ions, the fraction $F_c^3(\varepsilon)$ is linear versus the target thickness in the range studied ($\varepsilon \leq 8\times10^{14}$ at/cm2). Double collision processes are found negligeable and the single-collision conditions are fulfilled. The single electron capture cross section $\sigma_c^n$ is then deduced from the slope of the straight line (see Fig. 3).

A second method, the branching ratio method, has also been used to deduce the capture cross section from the data. From the spectra of the detection of the neutral fragments, as the ones shown in Figure 2, we determine the branching ratio $R_{c/N}^n$ between the number of electron capture events, peak $S_N = n$, over the total number of events in the total spectrum. The branching ratio is calculated after substraction of the spurious events. Thus, for a given target thickness for which the single-collision conditions are fulfilled, we can write

$$R_{c/N}^{n} = [N_c^{n}(\varepsilon)-N_c^{n}(0)]/ [N_N^{n}(\varepsilon)-N_N^{n}(0)] \qquad (2)$$

where $N_c^n(\varepsilon)$ and $N_N^n(\varepsilon)$ are the number of electron capture events and the total number of events in all the spectrum, respectively, for a target thickness $\varepsilon$, and, $N_c^n(0)$ and $N_N^n(0)$ are those measured with no helium target. Besides, we measure by means of the growth-rate method the cross section $\sigma_N^n$ of the production of at least one neutral fragment in the cluster fragmentation [15]. Then, $\sigma_c^n$ is given by $\sigma_c^n = R_{c/N}^n\ \sigma_N^n$.

For the $H_3^+$ case discussed above, we obtain by this method a value for $\sigma_c^n$ equal to $(3.8 \pm 0.4)\times10^{-17}$ cm$^2$ which is in good agreement with the one deduced by the growth-rate method ($(4.0 \pm 0.4)\times10^{-17}$ cm$^2$).

## IV. RESULTS AND DISCUSSION

In Figure 4a are reported the electron capture cross sections as a function of the cluster size n. We also report the measured values for incident molecular ions, $H_2^+$ and $H_3^+$. The value for incident protons has been taken from literature [4]. All the results have been obtained for the same velocity of the projectile, 1.55 $v_o$. It should be noted that the results have been extracted from several sets of measurements corresponding to different run times. The results displayed in Figure 4a show the good reproductibility of the measurements. In Figure 4b the mean value of the cross section for a given size $\sigma_c^n$ is plotted versus the cluster size.

First, we observe a decrease of the electron capture cross section from $H^+$ to $H_3^+$Such a decrease of the electron capture has also been observed by Abraham et al.[16] with $D_2^+$ and $D_3^+$ molecular incident ions colliding with argon at 100 keV/u. Besides, in the present work, for the cluster ions, i.e. $n \geq 5$, the capture cross section is observed to be independent of the cluster size. The straight line plotted in Figure 4b corresponds to an mean value $< \sigma_c^n >$ of the cross sections over all the cluster sizes ($< \sigma_c^n > = (4.4 \pm 0.4)\times10^{-17}$ cm$^2$). The $< \sigma_c^n >$ value is very close to the cross section obtained with the incident $H_3^+$ ion $(3.9 \pm 0.4)\times10^{-17}$ cm$^2$.

Concerning the results on $H^+$, $H_2^+$ and $H_3^+$ ions, one can observe that the number of exit channels associated to the electron capture process increases with the ion size and could induce increasing curve crossing :

$$H^+ + He \longrightarrow H + He^+$$

$$\begin{aligned} H_2^+ + He &\longrightarrow H_2 + He^+ \\ &\longrightarrow H + H + He^+ \end{aligned}$$

$$\begin{aligned} H_3^+ + He &\longrightarrow H_3 + He^+ \\ &\longrightarrow H_2 + H + He^+ \\ &\longrightarrow H + H + H + He^+. \end{aligned}$$

In our case, the time during the collision is short compared to the typical time of the motion of the protons in the molecular ion and the results obtained with the $H_2^+$ and $H_3^+$ molecular ions could be compared to the proton case with respect to the charge localization. The charge is not localized on a single proton but distributed over all the protons of the molecular ion. The most probable distance d(H-H) between the protons in the $H_2^+$ ions delivered by the accelerators (d(H-H) = 1.2 Å) [17] and the one in the $H_3^+$ molecular ions (d(H-H) = 1.1 Å) [18] could be compared with the largest impact parameter for capture by a proton, $R_c^1$. In a simple geometric model, $R_c^1$, the so-called capture distance, is deduced from the experimental value of the electron capture cross section by protons (see Fig. 3 and ref. 5) ($8\ 10^{-17}\,cm^2$) as follows

$$R_c^1 = (\sigma_c^1 / \pi)^{1/2} = 0.5\ \text{Å}.$$

The most probable distance d(H-H) in the $H_2^+$ molecular ion is about twice as large as $R_c^1$ the capture distance deduced from the proton case. Yet, the electron of the projectile has to be taken into account and could screen the charge during the capture process. Moreover, due to the triangular structure of the $H_3^+$ ion, this screening effect could be stronger for $H_3^+$ than for $H_2^+$ . Such screening effects connected to the delocalization of the positive charge in the molecular ion could explain the decrease of the electron capture cross section observed in Figure 4b from $H^+$ to $H_3^+$. Further experiments especially at various velocities should allow more insights in this result.

Turning now to the cluster ions, one of the most important features is the fact that the electron capture cross section is found independent of the cluster size. No structure effect involving the geometric shells of $H_2$ seems to be present. Another important feature is the mean value of the capture cross sections of the clusters (see Figure 3b) which is nearly equal to the $H_3^+$ value.

Quantum chemical calculations have been focused on exploring the ground potential energy surface of small $H_n^+$ clusters [2]. Such studies uncovered that the minimum energy structures correspond to a cluster formed by a three-center-bonded $H_3^+$ core with bond lengths in the order of 0.87 Å, solvated by essentially unperturbed $H_2$ molecules at distances of typically 1.6 Å in the first shell and more than 2 Å in the second shell. Going now to the cluster ions delivered by the cluster facility, previous experiments on hydrogen cluster collisions with thin foils [19] allowed to show that the $H_3^+$ core and the $H_2$ molecules are in their fundamental states. We can notice that the distance between the protons is smaller in the $H_3^+$ core of the clusters (d(H-H) = 0.87 Å ) than the most probable distance for the $H_3^+$ ion (d(H-H) = 1.1 Å ).

The absence of structure effect for the smaller sizes, as for example the size 9 which is the first geometric shell of three molecules around the $H_3^+$ core, shows that the electron capture process requires collisions at close distance of the $H_3^+$ core ion. Roughly, we can estimate that electron capture occurs mainly below the impact parameter $R_c^n$ deduced from the mean value of $\sigma_c^n$ as follows :

$$R_c^n = (<\sigma_c^n>/\pi)^{1/2} = 0.4 \text{ Å}.$$

This value has to be compared to the distance between the component of the cluster. The distance between the $H_3^+$ core and the added $H_2$ molecules in $H_9^+$ cluster ( 1.6 Å) is four times larger than the maximum limit of the impact parameter $R_c^n$. Thus the capture process is localized near the $H_3^+$ core. Since the mean value of $\sigma_c^n$ is nearly equal to the value of $\sigma_c^3$, the process of electron capture by hydrogen clusters on helium atoms seems to be the electron capture by the $H_3^+$ core of the cluster. That confirms the localization of the charge on the $H_3^+$ core in the clusters as suggested by theoretical works [1,2]. Nevertheless, the small difference observed between the mean value of $\sigma_c^n$ and the value of $\sigma_c^3$ is not significant if we take into account the experimental uncertainties. But a difference could have been expected since the incident $H_3^+$ ions and the $H_3^+$ core of the incident-clusters are not in the same vibrational state as explained before.

Another point has to be emphasized. Even for bigger sizes, such as n=35, the capture cross section is observed to be independent of the cluster size. The electron capture process is not disturbed by the sixteen $H_2$ molecules of the $H_{35}^+$ cluster. In fact, a decrease of the cross section when increasing size could be expected for large sizes due to a geometric screening of the $H_3^+$ core by the $H_2$ molecules. Indeed, the $H_n^+$.- He collision should induce an ionization of

$H_2$ before an electron capture, for example. The absence of geometric screening even with sixteen $H_2$ molecules in the cluster is intriguing and should lead to consider a kind of rather tubular structure for the hydrogen clusters with the $H_2$ molecules organized around an axis perpendicular to the triangular $H_3^+$ core.

## IV. CONCLUSION

We have measured the electron capture cross section in collisions between mass-selected swift hydrogen clusters ions $H_n^+$ and atomic helium, in a large size range ( $3 \leq n \leq 35$ ) at a given velocity of 1.5 $v_0$ (60 keV/u).

The results obtained with the molecular ions show a decrease of the electron capture cross section from $H^+$ to $H_3^+$. Such behavior can be connected to the relatively close distance between the helium and the projectile during the collision and to the delocalization of the charge on the molecular ion.

For the hydrogen clusters, the electron capture cross section is independent of the cluster size. The mean value of the capture cross sections for clusters is nearly equal to the $H_3^+$ capture cross section. That shows that the electron capture by hydrogen clusters on a helium atom is a process involving mainly the $H_3^+$ core and confirms the localization of the charge of the cluster on the $H_3^+$ core. An intruiging result is the fact that the cross section of this very localized electron capture process is not disturbed by the presence of sixteen molecules around the $H_3^+$ core. These experimental results should deal to further theoretical investigations on the geometric shells of $H_2$ molecules in ionized hydrogen clusters. Further experiments with other targets such as molecular targets should allow new investigations on the size dependence of the electron capture by a cluster.

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## FIGURE CAPTIONS

**Figure 1 :** The schematic experimental set-up.

**Figure 2 :** **a)** Detection of the neutral fragments for a given number of $H_{13}^+$ incident clusters ($\approx$1.1 $10^5$) for a target thickness of $(1.41\times10^{14})$at/cm$^2$. In the spectrum, each peak corresponds to a value of the sum of the mass numbers for all the neutral fragments coming from one $H_{13}^+$ dissociated cluster ($S_N$).

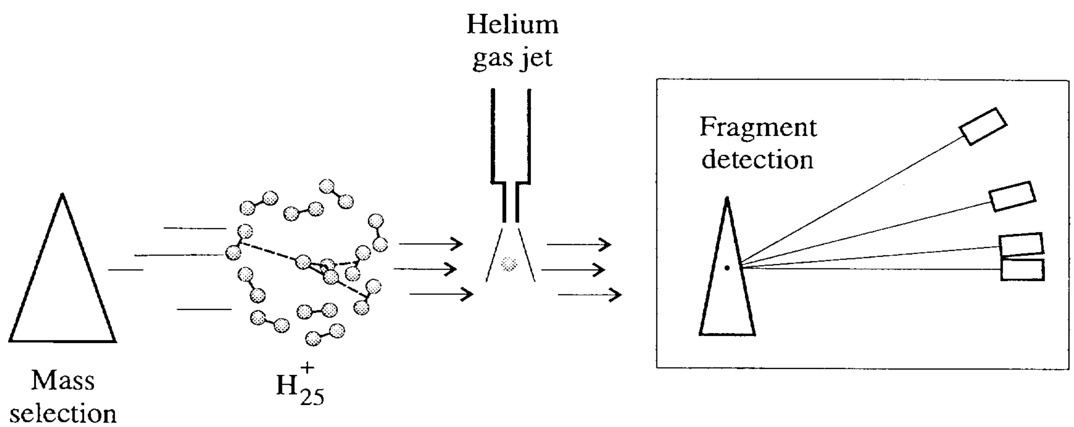
**b)** The same as a) without helium target.

**Figure 3 :** The fraction $F_c^n(\varepsilon)$ of electron capture events (see text) versus the target thickness $\varepsilon$ for incident $H_3^+$ ions.

**Figure 4 :** The electron capture cross section $\sigma_c^n$ as a function of the cluster size n ($1\leq n\leq35$). The value for the incident protons (■) has been taken from literature [5]. All the results have been obtained for the same velocity of the projectile, 1.55 $v_o$.

**a)** Various symbols correspond to different sets of measurement.

**b)**The capture cross section $\sigma_c^n$ (average value of the different experimental points for a given size) is plotted versus the cluster size n. The straight line corresponds to the mean value of all the cluster cross sections $<\sigma_c^n>$ , n=5 -35, odd,[ $(4.4\pm 0.4)\times10^{-17}$ cm$^2$ ].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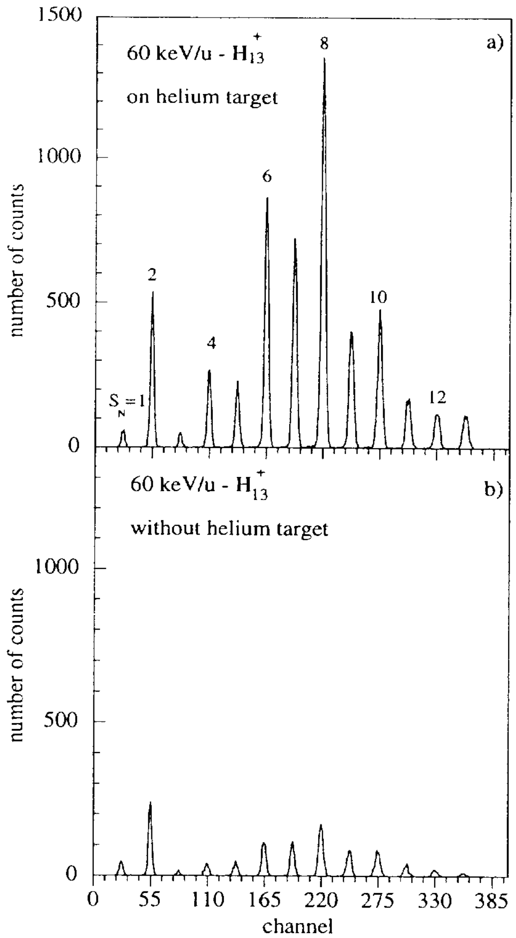


Figure 2 : Louc et al

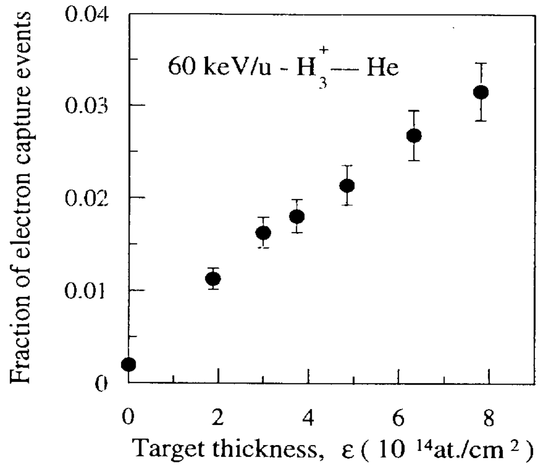


Figure 3 : Louc et al.

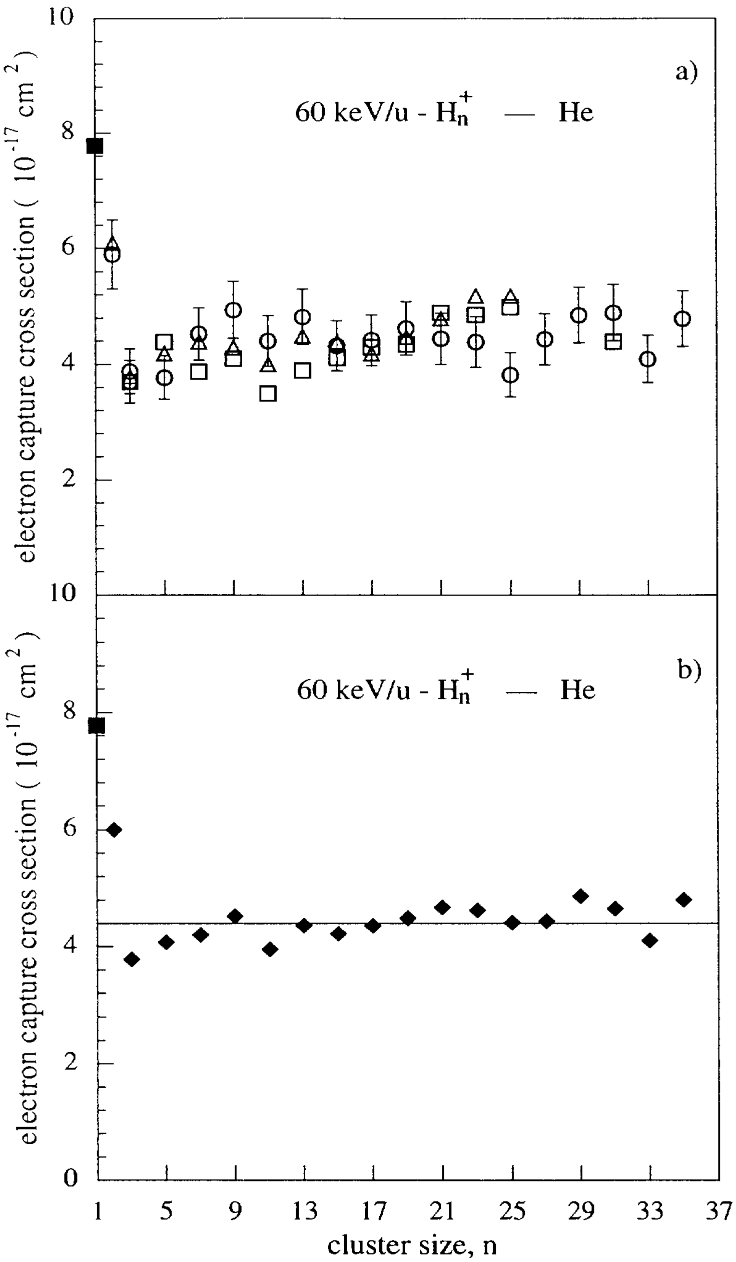


Figure 4 : Louc et al.